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(54) Title: FIBER GLASS BINDER COMPOSITIONS WITH REDUCED DUSTING

(57) Abstract: A resin binder composition useful in the manufacture of fiber glass articles is described. The resin binder is formulated to exhibit increased stress tolerance (flexibility) and to enable the manufacture of fiber glass products with less dusting with minimal compromise of physical strength characteristics.

FIBER GLASS BINDER COMPOSITIONS WITH REDUCED DUSTING

Field of the Invention

The present invention relates to an improved fiber glass composition.
5 More particularly the invention is directed to a fiber glass composition including an improved resin binder having enhanced flexibility and reduced dusting.

Background and Summary of the Invention

Fiber glass compositions find significant use as acoustical or thermal
10 insulation in a wide variety of commercial applications. Generally fiber glass compositions are prepared by applying a resin binder composition including a resole (phenol-formaldehyde) resin and a catalyst to glass fibers; the binder-coated glass fibers are collected in the form of a non-woven fiber glass blanket. The fiber glass blanket is thereafter compressed and heated to cure the binder composition either in
15 an oven ("line cure") to form batts, or in a mold to form compressed mats or other shaped fiber glass articles. During the heating step, the resin binder composition dispersed on the glass fibers polymerizes to form a cured resin solid that binds the glass fibers where they overlap to impart strength and resiliency to the fiber glass product. Fiber glass products formed using thermally cured resin binders thus retain
20 their shape due to resin crosslinking of the component fiber glass particles.

Fiber glass batts, mats, and other shaped fiber glass products are often combined with other construction materials to form insulated products. If the cured binder resin does not exhibit some threshold flexibility, the fiber glass product can readily lose structural integrity as a result of handling and manipulation in the
25 manufacture and installation of insulated products. Additionally, glass fiber dust can be generated during the handling and manipulation of the fiber glass product and during the useful life span of the final product, as the fiber glass product cracks and disintegrates under applied stress.

There have been significant research and development efforts directed
30 to improving resin binder compositions useful in the manufacture of fiber glass articles. The present invention is based on the development and use of novel catalyzed resole resin binder compositions which exhibit increased stress tolerance

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(flexibility) and concomitantly less friability and dusting without substantial compromise of physical strength characteristics.

The improved resin binder composition in accordance with this invention includes an aqueous solution comprising a resole resin, catalyst, and a
5 flexibilizer. The compositions include about 60% to about 90% by weight water and about 10% to about 40% by weight binder solids. The flexibilizer component(s) have polyhydric-functional and/or acid-functionality and typically form about 20 to about 50 percent per weight of the binder solids. The solid component of the binder composition optionally, but preferably, includes, an -NH₂ functional resole reactive
10 compound selected from the group consisting of urea, melamine, dicyandiamide, and methylol and C₁-C₄ alkoxymethyl derivatives thereof. The resin binder composition when used in the manufacture of thermally cured fiber glass compositions is characterized by improved flexibility and reduced dusting without substantial compromise of physical strength characteristics. The chemical/mechanical properties
15 of the improved resin binder composition allow preparation of fiber glass compositions exhibiting advantages in many applications.

In one embodiment of the present invention, the resin binder composition comprises an aqueous solution of resin solids comprising a resole resin, a catalyst, and both polyhydric-functional and acid-functional flexibilizers. The resin
20 solids comprise about 45 to 65 percent weight of the resin binder composition, while the flexibilizers comprise about 10 to about 25 percent by weight of solids each. The preferred acid-functional flexibilizers are water dispersible oils. Examples of water dispersible oils include maleinized unsaturated oils such as linseed-tung oil or soybean oils, sulfonated oils such as sulfonated fish oil or lard oil, and sulfited oils
25 such as bisulfited herring oil. Polyhydric flexibilizers are polyhydric alcohols, including di-, tri-, tetra-, and pentahydric alcohols having a molecular weight of less than 10,000, more preferably less than 5000, for example, Pentek (C(CH₂OH)₄), trimethylol propane, propoxylated trimethylolpropane, and propoxylated pentaerythritol.

30 In another embodiment, the flexibilizer consists essentially of one or more polyhydric-functional compounds forming about 30 to 45 percent of the weight of the resin solids. In still another embodiment, the flexibilizer component consists

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essentially of one or more acid-functional water dispersible oils forming about 15 to 35 percent of the resin solids.

Still another embodiment of the present invention is a process for preparing a fiber glass blanket characterized by improved flexibility and reduced
5 dusting. The method comprises the steps of forming an improved aqueous resin binder composition comprising a resole resin, a catalyst, and a flexibilizer in a total amount of 15 to 45% of the resin solids, coating glass fibers with said improved resin binder compositions, collecting the coated glass fibers to provide the fiber glass composition in the form of a non-woven mat containing up to about 25% by weight of
10 the resin binder composition on a solids basis, and drying the mat to a water content of less than 8% by weight. The process can further include the step of compressing the fiber glass blanket for storage and/or molding and curing the blanket for final use.

The solids of the present binder compositions also include a catalyst, and optionally may include an $-NH_2$ functional resole reactive compound, a silane in
15 an amount effective to decrease resin hydrophilicity, and/or a surfactant to aid in solubilizing intermediates and maintaining the binder as a homogenous solution.

Additional features of the present invention will become apparent to those skilled in the art upon consideration of the following detailed description of preferred embodiments exemplifying the best mode of carrying out the invention as
20 presently perceived.

Detailed Description of the Invention

The present invention is directed to improved fiber glass compositions and to methods of preparation of such compositions using an improved resin binder
25 composition, including a resole resin, a catalyst, and a polyhydric-functional and/or acid-functional flexibilizer. The composition may also include one or more resin binder modifiers, preferably urea, but other amines, such as melamine, and dicyandiamide, and methylol and $C_1 - C_4$ alkoxymethyl derivatives thereof may be suitable. The acid-functional flexibilizers include water dispersible oils, such as
30 maleinized, sulfonated oils, and sulfited oils while the polyhydric-functional flexibilizers include di-, tri-, tetra-, and penta-hydric alcohols having a molecular weight of less than 10,000, more preferably less than 5000. In preferred embodiments

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the catalyst for the resin binder composition consists of an ammonium toluene sulfonic acid ("TSA") solution, but other catalysts are known and may be used in accordance with this invention. See, for example, U.S. Patent No. 5,243,015. It has been found that the use of flexibilizers, as taught in this invention, are useful in the
5 manufacture of flexible fiber glass compositions.

Generally in the manufacture of the present fiber glass compositions an aqueous resin binder composition having about 10% to about 40%, more typically about 12% to about 35% solids is applied to hot glass fibers which are collected in the form of a non-woven blanket. The blanket is typically compressed and heat-cured
10 either in a line oven or in a heated compression mold; or it is rolled and stored for later heat-cure processing. The amount of resin binder composition used in the manufacture of the fiber glass blankets depends significantly on the intended use of the fiber glass product. Thus the binder can constitute about 3% up to about 25% by weight on a solids basis of the uncured fiber glass blanket. More typically the amount
15 of resin binder ranges from about 3% to about 20% by weight of the fiber glass blanket. Fiber glass blankets intended to be used in the manufacture of fiber glass batts can include as little as 3% resin binder. Fiber glass blankets intended for the manufacture of pipe insulation typically includes less than 10% by weight binder on a solids basis, more preferably less than about 8% by weight. Fiber glass compositions
20 in accordance with this invention may be used as automobile head liners, wherein the composition may include approximately 15 to 20% of resin binder.

Fiber glass blankets manufactured in accordance with this invention, as mentioned above, are typically compressed and heat-cured either in a line oven to form batts or boards or in compression molds to form molded fiber glass constructs.
25 Typically the fiber glass blankets are heated during that process to about 300° to about 500°F (about 149° to about 260°C), more typically about 400°F (about 204°C), for a period of time sufficient to cure the binder component of the fiber glass blanket. It has been found that preferred embodiments of the present improved resin binder composition including a polyhydric-functional and/or acid-functional
30 flexibilizer exhibit improved flexibility and less dusting. Such fiber glass compositions can withstand increased bending with less breakage than similar compositions lacking the flexibilizer. The resin binder portion of such fiber glass

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compositions is less friable and exhibits less dusting. Thus, products made from the fiber glass compositions of this invention have a longer useful life span. Moreover, installation of fiber glass components, such as automobile head liners, may take place with fewer fiber glass articles wasted due to breakage.

5 The resin binder compositions of the present invention include a resole resin, a flexibilizer, a catalyst, and optional binder modifiers. The resole resin is a phenol-aldehyde resin having a molar ratio of phenol to aldehyde of about 1:1.1 to about 1:5. Preferably the phenol to aldehyde ratio is from about 1:2 to about 1:3. Resole resins are commercially available from numerous resin suppliers. One
10 commercially available resin that can be used for formulating the present improved binder composition is GP® 454T81 ("T81"), Georgia-Pacific Resins, Inc., Decatur, Georgia.

 The phenol component of the resole resin can include a variety of substituted and unsubstituted phenolic compounds. The aldehyde component of the
15 resole resin is preferably formaldehyde but can include so-called masked aldehydes or aldehyde equivalents such as acetals or hemiacetals. Specific examples of suitable aldehydes include: formaldehyde, acetaldehyde, propionaldehyde, butyraldehyde, furfuraldehyde, and benzaldehyde.

 Flexibilizers are added to the resin binder composition to improve
20 flexibility and reduce dusting without sacrificing strength of the fiber glass composition. The flexibilizers of this invention include polyhydric-functional and acid-functional compounds. Preferably, the polyhydric-functional and acid-functional flexibilizers are present in about a 1:1 ratio and together comprise about 20 to 50 percent weight of the resin solids, most preferably about 40 percent weight of the
25 solids. However, the ratio of polyhydric-functional flexibilizer to acid-functional flexibilizer may vary greatly, to the point that polyhydric-functional flexibilizers may be used in the absence of acid-functional flexibilizers and acid-functional flexibilizers may be used without polyhydric-functional flexibilizers.

 The acid-functional flexibilizers of this invention are water dispersible
30 oils. Preferred flexibilizers are maleinized oils, including maleinized linseed-tung oil and maleinized soybean oils. Examples of other water dispersible oils include sulfonated oils such as fish oils and lard oils, and sulfited oils such as bisulfited

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herring oils. Many such oils are available commercially. In some applications it may be preferable to use purified oils, especially for those applications in which odor may be a problem. Polyhydric flexibilizers are polyhydric alcohols, including di-, tri-, tetra-, and pentahydric alcohols having a molecular weight of less than 10,000, preferably less than 5,000. Examples of polyhydric-flexibilizers include Pentek (C(CH₂OH)₄), trimethylol propane, propoxylated trimethylolpropane, and propoxylated pentaerythritol. Polyhydric-functional flexibilizers are reactive in that they participate in the polymerization of the resin matrix and alter its structure. The acid-functional flexibilizers are also capable of participating in the polymerization reaction. However, the acid-functional flexibilizers may also impart flexibility in a non-reactive manner by acting as plasticizers.

The catalyst component of the resole resin binder composition of this invention comprises an amine salt of a strong acid, preferably a sulfonic acid. The nature/structure of the sulfonic acid is not critical provided that it is soluble in the aqueous resin binder composition and provided its acid functionality is capable of catalyzing polymerization of the resole resin. A preferred catalyst may be TSA. Other preferred catalysts may be ammonium sulfate or an ammonium salt of an aryl sulfonic acid.

The catalyst is typically used in an amount ranging from about 3 to about 15 parts per hundred of the solid content in the resin binder composition. Preferably, the catalyst is present in an amount from about 10 to about 12 parts per hundred on a solids basis. The solids content is the amount of solid residue that remains after all volatile components of the binder composition are removed. The percentage of solids content in the binder is determined by placing about 1.0 gram of the binder composition in an aluminum dish and drying the composition to dryness in a circulating oven at 300°F (149°C). The weight of the dried composition is divided by the weight of the aqueous binder then multiplied by 100 to obtain the percentage of solids content in the binder.

The resin binder composition can be formulated to contain up to about 20% by weight on a solids basis of a binder modifier. Preferred binder modifiers are urea and silanes. Preferably, urea may be used in amounts up to about 50% of the resin solids, with 10% being most preferable. Urea is typically added to act as a

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formaldehyde scavenger and a binder extender. Urea readily combines with the free aldehyde in the resin under mildly alkaline or neutral pH conditions. It is important to reduce the free aldehyde content because aldehyde emissions, either during the curing process or later from the cured fiber glass article, are undesirable. The addition of

5 urea to the present resin binder composition reduces aldehyde emissions, improves the efficiency of resin cure and ultimately reduces the cost of the resin binder composition. Silanes, such as Silquest A1101, may be used in small amounts, preferably less than 1%, and most preferably about 0.2%. These modifiers impart thermal stability to the cured fiber glass product and they work to otherwise improve

10 binder performance. Silanes also decrease the hydrophilicity of the resin, thereby reducing the ability of the fiber glass product to attract moisture.

Surfactants may also be added to the resin binder composition to help solubilize intermediates and products formed from the condensation reaction between the phenol, the aldehyde and other binder additives. In addition, surfactants or

15 additives help disperse the condensation products and prevent agglomeration of these products to provide or maintain the binder as a homogeneous solution. Suitable surfactants include but are not limited to glycols, glycol ethers or esters, methoxylated alcohols, alkyl sulfonates and alkyl phosphonates.

The molded fiber glass compositions prepared using the improved

20 resins of this invention are expected to exhibit acceptable structural characteristics with improved flexibility and reduced dusting. Flexural, tensile, and compressive strengths of the molded fiber glass articles may be evaluated according to ASTM standard testing protocol D 638-91 "Standard Text Methods for Tensile Properties of Plastics" and D 790-92 "Standard Test Methods for Flexural Properties of

25 Unreinforced and Reinforced Plastics in Electrical Insulating Materials," and C 165-95 "Standard Test Methods for Measuring Compressive Properties of Thermal Insulations," respectively. Dusting is evaluated according to the General Motors 9627P:R1:CPCKMT dust fallout procedure.

Thus, another embodiment of the present invention is a process for

30 preparing a fiber glass blanket comprising glass fibers and a resin binder composition, wherein said fiber glass blanket is characterized by improved flexibility and reduced dusting compared to similar fiber glass compositions lacking the flexibilizer and/or

reactive diluent. The method comprises the steps of forming an improved aqueous resin binder composition comprising a resole resin, a catalyst, and a flexibilizer in a total amount of 30 to 40% of the resin solids, coating glass fibers with said improved resin binder compositions, collecting the coated glass fibers to provide the fiber glass composition in the form of a non-woven mat containing up to about 25% by weight of the resin binder composition on a solids basis, and drying the mat to a water content of less than 8% by weight. The process can further include the step of compressing the fiber glass blanket for storage and/or molding and curing the blanket for final use.

10 Examples

The following examples are set forth to illustrate the principles and practices of the present invention to one skilled in the art. They are not intended to be restrictive but merely to be illustrative of the invention. Unless otherwise stated, all parts, percentages and ratios are on a weight basis. All water was passed through a zeolite filled column to remove calcium and magnesium salts from the water. Solids for the following examples were generated by placing 1 gram of resin mixture in a pan and drying the resin in a circulating oven for 30 minutes at 300°F (149°C). The dry residue is weighed to determine the weight of solids. The percentage of solids content was determined by multiplying the ratio of the weight of dried, cured resin to the weight of the resin by 100 to obtain the percentage of solids content in the binder.

Flexibility was determined by a pipe test employing a series of pipes of differing diameter. Starting with the pipe of largest diameter, the dry residue was placed against the exterior of the pipe. The resin passed this phase of the test if it does not crack or break when pressed flush against the exterior surface of the pipe. The test is repeated using pipes of decreasing diameter.

Preparation 1: Phenol-Formaldehyde-Urea Premix T81.

Phenol/formaldehyde premix T81 was made to have a phenol-formaldehyde molar ratio of 1:3, with about 4.5% free phenol and 1.8% free formaldehyde. Sufficient water was added to yield an aqueous resole resin solution mixture having about 45% solids content. An aqueous solution of urea (40% by

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weight of urea) was added T81 premix in a ratio of 9:1 T81 premix to urea solution (by weight), while the resulting solution was stirred to ensure homogeneous mixing.

Preparation 2: Phenol-Formaldehyde-Urea Premix K-3-2A.

- 5 Phenol and formaldehyde in a phenol-formaldehyde molar ratio of 1:3.2 were reacted together to obtain a resole resin having about 1% free phenol and 7% free formaldehyde. Sufficient water was added to yield an aqueous resole resin solution mixture having about 51% solids content. Phenol-formaldehyde resin (6263 pounds), to which an aqueous solution of urea (3422 pound, 40% by weight of urea)
- 10 was added, while the resulting solution was stirred to ensure homogeneous mixing. The temperature of the resulting solution was maintained from about 50°F to about 70°F (10°C to about 21°C). This premix solution had a density of about 9.69 lb/gallon and a solids content of about 47.1±2%.

15 Example 1: Preparation of a Resole Resin Binder Composition Containing Maleinized Linseed-Tung Oil

- To a stirred container 106.1 grams phenol-formaldehyde-urea premix (Preparation 2) was added. Next, a solution of 15.3 grams Pentek dissolved in 148.7
- 20 grams soft water at 116°F (47°C) was added. Temperature was reduced to 80°F (27°C), and 3.0 grams of ammonia were added. Eight minutes later, the ammonia was followed by the addition of 30.0 grams of maleinized linseed-tung oil (Ohio Polychemicals, Polychem 7238-100), and the temperature was reduced to 75°F (24°C). 30 grams 15% ammonium TSA solution and 0.2 grams of Silquest A1101
- 25 silane were added and stirred for 15 minutes. The resulting resin mixture was dried in a circulating oven for 30 minutes at 300°F (149°C). The pipe test showed some flexibility, but the material was too weak to provide good test results.

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Example 2: Preparation of a Resole Resin Binder Composition Containing Maleinized Linseed-Tung Oil using Preparation 1

Preparation 1 was formed using 100.0 grams of T81 and 12.5 grams of urea solution. A Pentek solution was prepared by adding 19.8 grams of Pentek to 113.9 grams of soft water at about 160°F (71°C), to dissolve all of the Pentek. The Pentek solution was added to the mix, and the mix was 146°F (63°C). Five minutes later, 0.29 grams of ammonia were added, followed nine minutes later by 20.0 grams of maleinized linseed-tung oil (as used in Example 1). A few minutes later, 66.7 grams 15% ammonium TSA solution and 0.2 grams A1101 silane were added. The mixture was baked for 30 minutes at 300°F (149°C). The bakeout was yellow in color and could be flexed without breaking. The bakeout of this example passed the 1/4 inch (6.4 mm) pipe test.

A comparable but larger-scale preparation was used to test dusting. An initial dust test according to the General Motors 9627P:R1:CPCCKMT dust fallout procedure showed 49.9 mg of dust collected for the sample prepared with maleinized linseed-tung oil, as compared to 71.0 grams from the standard uncured amber resin and 95.5 to the standard uncured black resin. Thus, as compared to similar standard resins, the resins of the present invention may exhibit reduced dusting.

20

Example 3: Preparation of a Resole Resin Binder Composition Containing Sulfonated Fish Oil

The phenol/formaldehyde resin of Example 2 was prepared, except that 20.0 grams of sulfonated, oxidized fish oil (Atlas Refinery Inc., Eureka 400-R) was substituted for the maleinized linseed-tung oil of Example 2. The resulting bakeout was tan in color, and it passed the 1/4 inch (6.4 mm) pipe test.

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Example 4: Preparation of a Resole Resin Binder Composition Containing Sulfonated Lard Oil

The phenol/formaldehyde resin of Example 2 was prepared, except that
5 20.0 grams of sulfonated, oxidized lard oil (Atlas Refinery Inc., Eureka 800-R) was substituted for the maleinized linseed-tung oil of Example 2. The resulting bakeout was tan in color. It passed the 1/2 inch (12.7 mm) pipe test, but failed the 1/4 inch (6.4 mm) pipe test.

10 Example 5: Preparation of a Resole Resin Binder Composition Containing Bisulfited Herring Oil

The phenol/formaldehyde resin of Example 2 was prepared, except that
20.0 grams of polymerized oxidized bisulfited herring oil (Böhme-Filtrex Cutapol
15 TIS-MF3) was substituted for the maleinized linseed-tung oil of Example 2. The resulting bakeout was tan in color. The bakeout of this example passed the 2 inch (50.8 mm) pipe test but failed the 1 inch (25.4 mm) pipe test.

CLAIMS:

1. A resin binder composition comprising an aqueous solution or dispersion of binder solids comprising a resole resin, a catalyst, and a flexibilizer
5 selected from the group consisting of polyhydric alcohols and acid-functional water dispersible oils.
2. The resin binder composition of claim 1 further comprising an -NH₂ functional resole resin reactive compound selected from the group of urea, melamine, dicyandiamide, and methylol and C₁-C₄ alkoxymethyl derivatives thereof.
- 10 3. The resin binder composition of claim 1 wherein the flexibilizer component forms about 20 to about 50 percent of the resin solids.
4. The resin binder composition of claim 3 wherein the flexibilizer component comprises a polyhydric alcohol and an acid-functional water dispersible oil.
- 15 5. The resin binder composition of claim 4 wherein the polyhydric alcohol and the acid-functional water dispersible oil are present in about a 1:1 ratio by weight.
6. The resin binder composition of claim 1 wherein the flexibilizer component is a maleinized oil.
- 20 7. The resin binder composition of claim 1 wherein the resin binder solids include about 45 to about 65 percent weight of the resole resin.
8. The resin binder composition of claim 1 further comprising a silane in an amount effective to decrease resin hydrophilicity.
9. The resin binder composition of claim 1 further comprising a
25 surfactant in an amount effective to solubilize and disperse the binder solids.
10. A fiber glass composition comprising a non-woven mat of glass fibers wherein the glass fibers are coated with the resin binder composition of claim 1.
11. The fiber glass composition of claim 10 wherein the binder composition further comprises an -NH₂ functional resole resin reactive compound
30 selected from the group of urea, melamine, dicyandiamide, and methylol and C₁-C₄ alkoxymethyl derivatives thereof.

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12. The fiber glass composition of claim 10 wherein the flexibilizer component comprises a polyhydric alcohol and an acid-functional water dispersible oil.

13. The fiber glass composition of claim 10 wherein the flexibilizer
5 component comprises a maleinized oil.

14. A method for preparing a fiber glass composition comprising
the steps of
coating glass fibers with the binder composition of claim 1,
collecting the coated glass fibers in the form of a non-woven mat, and
10 drying the mat to a water content of less than 8% by weight.

15. A method for preparing a fiber glass composition exhibiting
good flexibility and tensile strength and low dust production comprising the steps of
coating glass fibers with the binder composition of claim 1,
collecting the coated glass fibers in the form of a non-woven mat,
15 drying the mat to a water content of less than 8% by weight, and
compressing and heating the mat to a temperature sufficient to cure the
dried resin binder composition.

INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER IPC 7 C08L61/06 C08L61/14		Internat. Application No PCT/US 00/40611
According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) IPC 7 C08L		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practical, search terms used) EPO-Internal, WPI Data, PAJ		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X,P	WO 99 60043 A (HELBING CLARENCE ;KNAUF FIBER GLASS GMBH (US)) 25 November 1999 (1999-11-25) abstract page 3, line 28 -page 10, line 23; claims; examples page 7, line 11-18	1,2,7,8, 10,11, 14,15
X	US 3 288 734 A (OWENS-CORNING FIBERGLAS) 29 November 1966 (1966-11-29) column 2, line 16-50; claims; examples	1,10,14, 15
X	GB 961 029 A (THE DISTILLERS COMPANY.) 22 February 1961 (1961-02-22) examples 1-5	1,6
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<input checked="" type="checkbox"/> Further documents are listed in the continuation of box C. <input checked="" type="checkbox"/> Patent family members are listed in annex.		
* Special categories of cited documents : <div style="display: flex; justify-content: space-between;"> <div style="width: 45%;"> <p>*A* document defining the general state of the art which is not considered to be of particular relevance</p> <p>*E* earlier document but published on or after the international filing date</p> <p>*L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>*O* document referring to an oral disclosure, use, exhibition or other means</p> <p>*P* document published prior to the international filing date but later than the priority date claimed</p> </div> <div style="width: 45%;"> <p>*T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>*X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>*Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.</p> <p>*8* document member of the same patent family</p> </div> </div>		
Date of the actual completion of the international search <div style="text-align: center; font-weight: bold;">28 November 2000</div>		Date of mailing of the international search report <div style="text-align: center; font-weight: bold;">05/12/2000</div>
Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016		Authorized officer <div style="text-align: center; font-weight: bold;">Hutton, D</div>

INTERNATIONAL SEARCH REPORT

Internat. Application No

PCT/US 00/40611

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 819 441 A (FARGO H ET AL) 25 June 1974 (1974-06-25) column 2, line 15 -column 3, line 21; examples; table 1 ---	1,2,10, 11,14,15
X	US 5 708 121 A (ANDERSON KURT FORREST ET AL) 13 January 1998 (1998-01-13) abstract column 1, line 18 -column 7, line 55; claims; examples 1,2 ---	1,2,9-11
X	US 5 473 012 A (JOACHIM JACKY ET AL) 5 December 1995 (1995-12-05) column 6, line 58; claims; examples column 1, line 14 -column 7, line 43 ---	1,10
X	US 3 300 427 A (THE DOW CHEMICAL COMPANY) 24 January 1967 (1967-01-24) example 6 ---	1
X	US 4 324 833 A (YAU BEN J) 13 April 1982 (1982-04-13) the whole document ---	1,2,10, 11,14,15
X	US 4 210 562 A (MCCOMBS FRANK P) 1 July 1980 (1980-07-01) column 1, line 5 -column 6, line 5; claims; example ---	1,2,8, 10,11, 14,15
X	GB 1 155 004 A (MONSANTO COMPANY) 11 June 1969 (1969-06-11) claims; examples ---	1
A	EP 0 913 413 A (BORDEN INC) 6 May 1999 (1999-05-06) the whole document ---	1-15
A	US 4 025 473 A (RIFI MAHMOUD RASHAD) 24 May 1977 (1977-05-24) column 1, line 5 -column 5, line 64; example 3 ---	1-9
A	US 4 495 329 A (BLAIR ROBERT E ET AL) 22 January 1985 (1985-01-22) column 2, line 5 -column 4, line 68; examples -----	1,10

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 00/40611

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9960043 A	25-11-1999	NONE	
US 3288734 A	29-11-1966	NONE	
GB 961029 A		FR 1284228 A	02-07-1962
US 3819441 A	25-06-1974	AR 203625 A	30-09-1975
		AT 326289 B	10-12-1975
		AT 593973 A	15-02-1975
		AU 470890 B	01-04-1976
		AU 5771073 A	09-01-1975
		BE 801816 A	02-01-1974
		CA 1009458 A	03-05-1977
		DE 2334263 A	31-01-1974
		ES 416574 A	01-04-1976
		FR 2236804 A	07-02-1975
		GB 1382963 A	05-02-1975
		IT 1046495 B	30-06-1980
		JP 49058165 A	05-06-1974
		LU 67940 A	18-01-1974
		NL 7309440 A	08-01-1974
		ZA 7304486 A	28-08-1974
US 5708121 A	13-01-1998	CA 2143601 A	23-08-1996
US 5473012 A	05-12-1995	US 5300562 A	05-04-1994
		AT 181904 T	15-07-1999
		AU 655257 B	08-12-1994
		AU 1605192 A	12-11-1992
		CA 2068242 A	10-11-1992
		CN 1068585 A, B	03-02-1993
		CS 9201326 A	18-11-1992
		DE 69229523 D	12-08-1999
		DK 512908 T	31-01-2000
		EP 0512908 A	11-11-1992
		ES 2136075 T	16-11-1999
		FI 922104 A	10-11-1992
		HU 65113 A, B	28-04-1994
		JP 5178952 A	20-07-1993
		KR 248691 B	01-04-2000
		NO 921753 A	10-11-1992
		NZ 242678 A	27-04-1995
		PL 171328 B	30-04-1997
		SI 9200074 A	27-11-1992
		TR 27281 A	26-12-1994
		BR 9201753 A	29-12-1992
		ZA 9203160 A	27-01-1993
US 3300427 A	24-01-1967	NONE	
US 4324833 A	13-04-1982	NONE	
US 4210562 A	01-07-1980	NONE	
GB 1155004 A	11-06-1969	NONE	
EP 0913413 A	06-05-1999	US 5952440 A	14-09-1999
		AU 9043498 A	27-05-1999

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/US 00/40611

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
EP 0913413 A		JP 11172073 A NO 985092 A NZ 331562 A	29-06-1999 04-05-1999 23-12-1998
US 4025473 A	24-05-1977	CA 1068837 A	24-12-1979
US 4495329 A	22-01-1985	NONE	